

Understanding the Nature of Marine Aerosols and Their Effects in the Coupled Ocean-Atmosphere System

Armin Sorooshian, Principal Investigator

University of Arizona

PO BOX 210011

Tucson, Arizona 85721

phone: (520) 626-5858 fax: (520) 621-6048 email: armin@email.arizona.edu

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LONG-TERM GOALS

The long-term goal of this work is to understand the sources and nature of marine aerosol particles and how they influence visibility, cloud properties, the thermodynamic structure of the marine boundary layer, and the transmission of radiation.

OBJECTIVES

The objectives of this project extend across three areas: (i) advancing aerosol measurement techniques via the development of new instrumentation to quantify aerosol-water interactions and drop residual particle properties; (ii) improving knowledge and model predictions related to the physicochemical nature of aerosol particles and ocean-aerosol-cloud-precipitation-radiation interactions; and (iii) strengthening a research methodology leveraging multiple complementary tools of analysis to guide future studies of this nature in the marine atmosphere.

APPROACH

The main technical approach is to use a combination of in-situ aircraft measurements, cloud models, and satellite remote sensing data to study the nature and character of aerosols and their effects in the marine atmosphere over a broad range of spatial and temporal scales. The work during the last year included the following tasks:

- Characterize and conduct airborne measurements with a state-of-the-art instrument to quantify aerosol-water interactions, termed the differential aerosol sizing and hygroscopicity spectrometer probe (DASH-SP).
- Develop, characterize, and conduct airborne measurements with a new aircraft inlet, called the counterflow virtual impactor (CVI), to preferentially sample only drop residual particles when flying in clouds. This work was possible because of an ONR DURIP award (N00014-11-1-0783). This new inlet directly complements the research objectives associated with the ONR YIP award (N00014-10-1-0811).
- Use satellite remote sensing observations to study aerosol-cloud-precipitation-radiation interactions in the marine atmosphere.

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- Use cloud models to examine the salient features of cloud drop activation and the subsequent growth of cloud drops to precipitation-sized drops over a wide range of conditions.
- Carry out in-situ measurements in the marine atmosphere to simultaneously characterize aerosol physicochemical properties and to quantify the interactive nature between aerosol particles, oceans, clouds, precipitation, meteorology, and radiation.

Key scientists involved with this work include Dr. Fred Brechtel (Brechtel Manufacturing Inc.), who collaborated with the PI on the development of the two new instruments (DASH-SP and CVI). Dr. Graham Feingold (National Oceanic and Atmospheric Administration) collaborated with the PI on using cloud models and interpreting satellite data to study cloud behavior in the marine atmosphere. The PI collaborated with Drs. Haflidi Jonsson (Naval Postgraduate School), John Seinfeld (California Institute of Technology), Bruce Albrecht (University of Miami), Lynn Russell (UC-San Diego), and Athanasios Nenes (Georgia Tech) to conduct a combined ship-aircraft field study between July-August 2011 called the Eastern Pacific Emitted Aerosol Cloud Experiment (E-PEACE). An on-going collaboration with Dr. Daniel Partridge (University of Oxford) has involved conducting inverse modelling of aerosol-cloud interactions jointly with the use of airborne data for aerosol and cloud parameters in the marine atmosphere.

WORK COMPLETED

During FY2012, the main tasks that were completed included the following:

- Characterization and airborne deployment of the CVI and DASH-SP instruments
- Execution of the Eastern Pacific Emitted Aerosol Cloud Experiment (E-PEACE)
- Satellite data analysis and modeling studies to examine aerosol-cloud-precipitation interactions

Technical accomplishments in the previous year, include the following:

- Successful deployment and collection of unique aerosol data with the newly-developed CVI and DASH-SP instruments. Three peer-reviewed manuscripts have used CVI data in the past year and more are in development. A manuscript is in preparation to describe the new DASH-SP and data collected with it during an airborne experiment. These two instruments each provide a new capability with regard to studying the role of aerosol particles in the marine atmosphere.
- The two-month E-PEACE field experiment was the first of its kind in that there was an instrumented ship and airplane coordinating tracks to investigate how aerosols emitted from the ship impacted cloud properties, as monitored by the Center for Interdisciplinary Remotely Piloted Aircraft Studies (CIRPAS) Twin Otter. Four peer-reviewed manuscripts have resulted from the E-PEACE experiment and more are in preparation.
- Modeling, satellite data analysis, and ship/aircraft measurements have been used synergistically in new ways to push forward the understanding of what factors control stratocumulus properties, including cloud albedo. Two peer-reviewed manuscripts have resulted from this effort and more are in preparation.

RESULTS

Two new capabilities were introduced in the form of new instruments that were fully characterized and deployed on different airplanes in the past year for research measurements. The introduction of these instruments is critical as they will provide new state-of-the-art capabilities for aircraft platforms aiming to study aerosol-cloud interactions in marine atmospheres. The first instrument, the counterflow virtual impactor (CVI) inlet (Figure 1), was deployed on the CIRPAS Twin Otter for approximately 140 flight hours during the 2011 E-PEACE field experiment. This inlet allows for preferential selection of cloud drops in clouds, which then get dried to leave drop residual particles that can be sampled and characterized by downstream aerosol instrumentation. Without this inlet, measurements in clouds through traditional sub-isokinetic inlets lack meaning as there is no confidence in what is being sampled (i.e. some combination of shattered drops and interstitial aerosols). The new CVI also addresses other shortcomings from previous aircraft field experiments in marine atmospheres: (i) negligible contamination issues associated with the inlet material of construction; (ii) a significant sample flow rate to downstream instruments ($\sim 15 \text{ L min}^{-1}$) that reduces the need for dilution; and (iii) a high level of accessibility to the probe interior for cleaning (Shingler et al., 2012). E-PEACE data downstream of this inlet have already helped unambiguously detect the influence of specific sources on cloud drops, including a smoke plume generated on board the Research Vessel (R/V) Point Sur. Droplet residual particle composition measurements downstream of the CVI indicated that the plume-influenced drops were nearly entirely organic ($\sim 90\%$ by mass) as compared to the background marine aerosol ($\sim 65\%$ by mass). Such measurements are scarce and will greatly benefit the scientific community's understanding of aerosol effects in the marine atmosphere.

The second newly-developed instrument is the differential aerosol sizing and hygroscopicity probe (DASH-SP), which is displayed in Figure 2. This instrument was specifically built to be compact and provide rapid measurements (\sim seconds) of size-resolved sub-saturated aerosol hygroscopicity. This critical parameter is needed to accurately quantify how particles interact with light (which reduces visibility) and how effective they will be in activating into cloud drops. The DASH-SP was deployed between May and June 2012 on a DC-8 aircraft as part of the Deep Convective Clouds and Chemistry (DC3) field study, which included one flight probing the marine boundary layer. Owing to the success of this instrument during DC3, the DASH-SP will be proposed to be deployed in future aircraft-based studies, in addition to being used in focused laboratory experiments to study aerosol thermodynamics associated with water-uptake.

The Eastern Pacific Emitted Aerosol Cloud Experiment (E-PEACE) 2011 was a targeted aircraft campaign with embedded modeling studies, using the CIRPAS Twin Otter aircraft and the R/V Point Sur between July and August 2011 off the central coast of California, with a full payload of instruments to measure particle and cloud number, mass, composition, and water-uptake distributions (Figure 3). E-PEACE used three emitted particle sources to separate particle-induced feedbacks from dynamical variability, namely (i) shipboard smoke generated particles with $0.05\text{-}1 \mu\text{m}$ diameters (which produced tracks measured by satellite and had drop composition characteristic of organic smoke), (ii) combustion particles from container ships with $0.05\text{-}0.2 \mu\text{m}$ diameters (which were measured in a variety of conditions with droplets containing both organic and sulfate components), and (iii) aircraft-based milled salt particles with $3\text{-}5 \mu\text{m}$ diameters. A description of our two-month experiment is summarized by Russell et al. (2012), with additional manuscripts discussing more specific findings obtained thus far. For example, an analysis was conducted for stratocumulus cloud albedo responses in ship tracks, based on in situ aircraft measurements and three years of satellite observations of nearly 600 individual ship tracks (Chen et al., 2012). We determined that the sign

(increase or decrease) and magnitude of the albedo response in ship tracks depends on factors such as cloud top height, free tropospheric moisture, and mesoscale cloud structure. In a closed cell structure (cloud cells ringed by a perimeter of clear air), approximately a third of ship tracks exhibited a reduction in albedo.

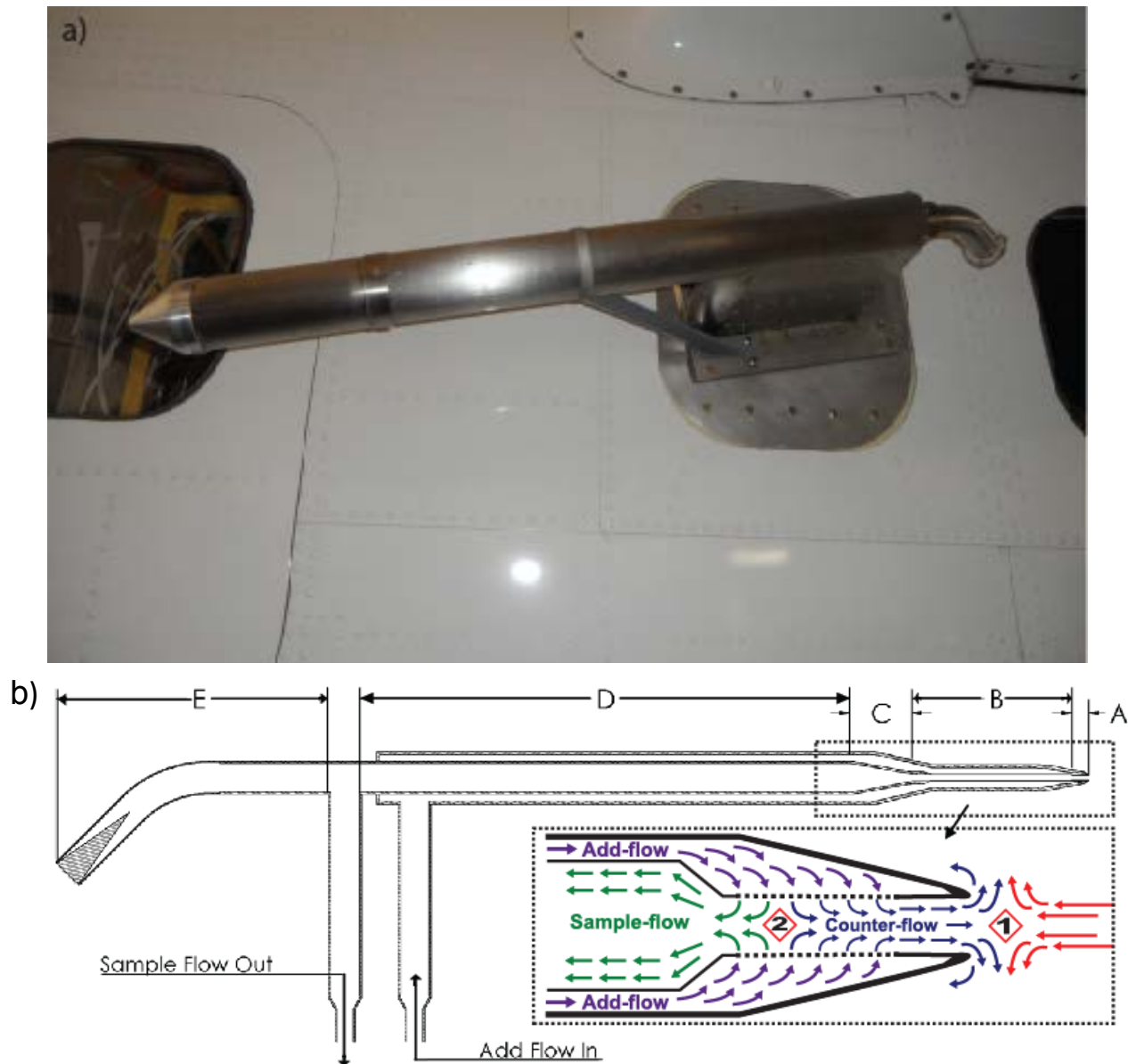


Figure 1. (a) Photograph of the newly-developed CVI inlet mounted to the left side of the CIRPAS Twin Otter during the 2011 E-PEACE field campaign. (b) Schematic depiction of the CVI inlet and the flows innate to its operation (Figure 1b adopted from Shingler et al., 2012). Sampled air enters the CVI through the inlet nozzle (denoted as A) and passes through a region containing a porous tube where heated counter-flow air is introduced. The resulting sample flow enters an extension tube (B), before the expansion region (C). The particles then travel through additional plumbing (D) and enter the aircraft body for sample feed to aerosol instrumentation. Entities too large to bend into the aircraft are collected in a particle trap (E). Two stagnation planes (labeled 1 and 2) are generated between opposing flow directions. The droplet cut size, which was $\sim 11 \mu\text{m}$ during E-PEACE, is governed by the velocity of the ambient air flow and the distance between the two stagnation planes.



Figure 2. Photograph of the newly-developed differential aerosol sizing and hygroscopicity probe (DASH-SP). The left compartment is the differential mobility analyzer (DMA), which selects dried particles of a given size, which are then transported to the right compartment. The latter compartment splits the monodisperse stream into two streams: one is kept dry and the other humidified to a pre-selected relative humidity between 5-95%. These two streams then enter custom-made optical particle counters to quantify the final wet size. The final parameter of interest is the “growth factor”, which is the ratio of the humidified particle diameter to the dry diameter.

E-PEACE provided an opportunity to examine the properties of marine aerosol and cloud water measured in the shipping lanes between Monterey Bay and San Francisco off the central California coast. Sub-cloud sub-micrometer aerosol particles exhibited average mass and number concentrations of $2 \mu\text{g m}^{-3}$ and 510 cm^{-3} , respectively, which are consistent with previous measurements in the same region. Enhancements in cloud water vanadium and cloud droplet number concentrations are observed concurrently with a decrease in cloud water pH, suggesting that periods of high aerosol loading are primarily linked to increased ship influence (Coggon et al., 2012). Mass spectra from a compact time-of-flight Aerodyne aerosol mass spectrometer revealed an enhancement in the fraction of specific organic mass spectral markers (i.e. m/z 42 and m/z 99) in ship-impacted clouds. These ions are well correlated with each other ($r^2 > 0.64$) both in and out of cloud and constitute 3-14% of organic mass during periods of enhanced sulfate. High-resolution mass spectral analysis of these organic masses from ship measurements suggests that the ions responsible for this variation were oxidized, possibly

due to cloud processing. Inter-relationships between numerous organic acids in cloud water (82 total samples collected) confirm documented aqueous-phase reaction mechanisms based on laboratory measurements, many of which are summarized in Figure 4. The E-PEACE chemical measurements are providing new insight into the role of aqueous-phase chemistry in modifying aerosol properties. Manuscripts are in preparation to summarize cloud water measurement results, including those associated with Figure 4.

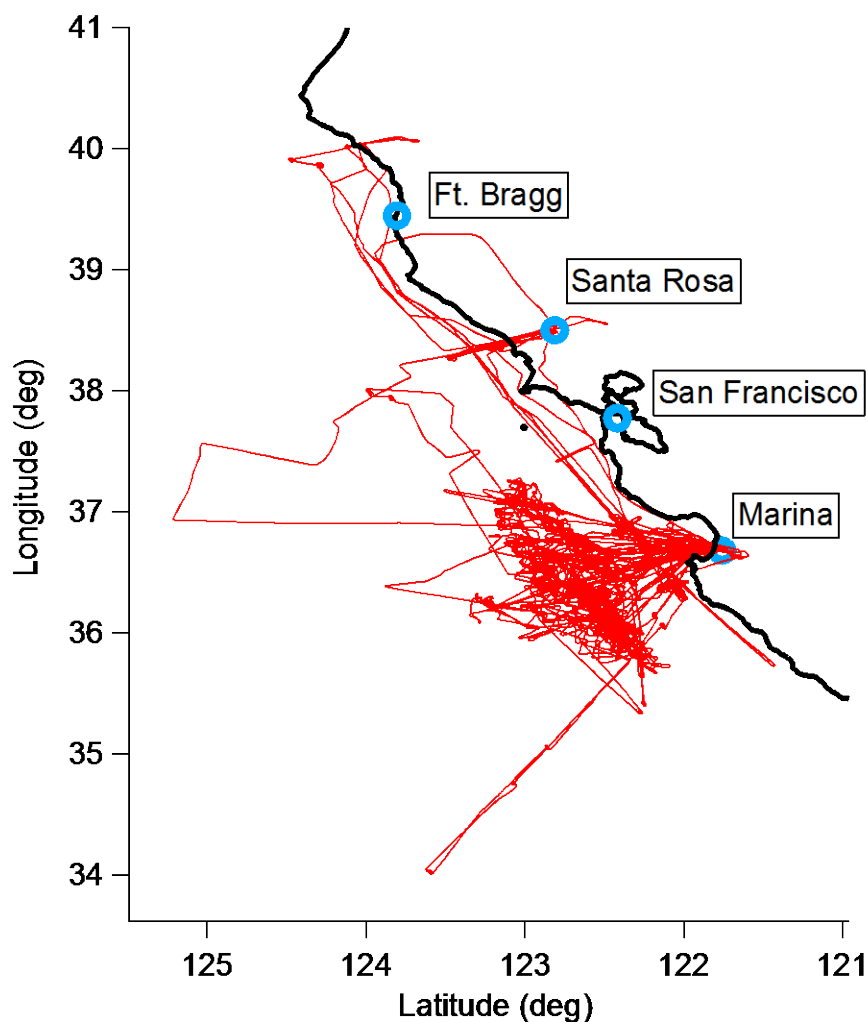


Figure 3. Spatial extent of 30 flights (in red) carried out by the CIRPAS Twin Otter during the 2011 E-PEACE field study. A central goal of this experiment was to examine how aerosol particle perturbations modify cloud microphysical and macrophysical properties. Most of the flights were focused on the region just south of the San Francisco area where there is intense ship traffic, which produces large aerosol perturbations that were ideal for the scientific objectives of E-PEACE.

In a collaborative effort, Partridge et al. (2012) introduced a novel approach to investigate aerosol-cloud interactions by coupling a Markov Chain Monte Carlo (MCMC) algorithm to an adiabatic cloud parcel model. While many numerical aerosol-cloud sensitivity studies have been carried out, few have used statistical analysis tools to investigate the global sensitivity of a cloud model to input aerosol physicochemical parameters. Using numerically generated cloud droplet number concentration (CDNC) distributions as cloud observations, our inverse modeling framework successfully estimates

the correct calibration parameters, and their underlying posterior probability distribution. Our work provides a new, integrative framework to evaluate the global sensitivity of the derived CDNC distribution to the input parameters describing the lognormal properties of accumulation-mode aerosol particles and their composition. Our analysis shows that the most important parameters in governing CDNC in typical marine conditions is the aerosol particle concentration and mean radius of the accumulation mode; however, in more polluted conditions (particle number concentration $> 400 \text{ cm}^{-3}$), the relative importance of the aerosol soluble mass fraction grows significantly. The competition and compensation between the cloud model input parameters illustrates that if the soluble mass fraction is reduced, the aerosol number concentration, geometric standard deviation and mean radius of the accumulation mode must increase in order to achieve the same CDNC distribution. This study demonstrates that inverse modeling provides a flexible, transparent and integrative method for efficiently exploring aerosol-cloud interactions with respect to parameter sensitivity. Several additional studies can now be carried out to extend this approach to examine other aspects of marine aerosol-cloud interactions.

Figure 4. A visual schematic of correlative relationships (r) between methanesulfonate, sulfate, and organic acids in cloud water samples collected during E-PEACE. The solid lines with arrows represent documented pathways, dashed lines with arrows represent a documented link with intermediate steps in between, and dashed lines with no arrows do not have a clear documented reaction pathway. Numbers signify statistically-significant correlation coefficients (r) at 95% confidence using a student's two-tailed t-test; underlined numbers are statistically insignificant. Since glyoxylate could not be quantified, but only detected in seven samples, the frequency of its coincident detection with other related species is reported.

IMPACT/APPLICATIONS

Both the CVI and DASH-SP are new instruments that are anticipated to find important use in subsequent field studies examining scientific aspects of marine aerosols and their effects on visibility, cloud properties, and the transmission of solar radiation.

TRANSITIONS

The DASH-SP (funded through a YIP award) and CVI (funded through a DURIP award) are instruments that have reached maturation. The small business that led the construction of the instruments, Brechtel Manufacturing Inc., is manufacturing more of these instruments for more widespread application.

RELATED PROJECTS

A related project is sponsored by the National Science Foundation and included support to assist with the PI's involvement with E-PEACE. This project specifically supported aerosol composition measurements on the R/V Point Sur and satellite data analysis in support of that field project.

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PUBLICATIONS

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